Reports

Energy Conservation Through More Efficient Lighting

Abstract. *The efficiency of a mercury-rare gas electrical discharge, which forms the basis of a fluorescent lamp, can be increased about 5 percent simply by increasing the concentration of mercury-196 from 0.146 percent (natural) to about 3 percent. These findings can be implemented immediately without any significant change in the process of manufacturing of this widely used source of illumination, provided that mercury-196 can be obtained economically. The potential energy savings for the United States are estimated to be worth in excess of $200 million per year.*

A weakly ionized plasma of mercury and rare gases under low pressure ($\sim 6 \times 10^{-3}$ and 1 to 3 torr, respectively) forms the basis of the fluorescent lamp. Electrical energy is converted to mercury resonance radiation (wavelength, 253.7 nm) at an efficiency of 55 to 65 percent (*1, 2*). This radiation in turn is converted to visible light by a solid phosphor. A limiting factor in the efficiency is that the 253.7-nm resonance radiation emitted from excited mercury atoms in the plasma is absorbed and reemitted many times by neutral mercury atoms during its escape to the walls of the discharge tube. This trapping of resonance radiation prolongs the effective lifetime of the excited atoms and increases the opportunity for radiationless energy conversions, which reduce efficiency.

We recently discovered (*3–5*) a simple way of decreasing resonance trapping in the mercury–rare gas plasma. The method may be very practical because of developments in isotope separation technologies over the past 10 years (*5*).

The 253.7-nm resonance line of mercury is composed of five hyperfine components, principally the result of isotope shifting (*6*). Under the electrical discharge conditions of a standard fluorescent lamp, the line shape is as shown in Fig. 1. The primary broadening mechanisms of the hyperfine components are Doppler and Lorentz broadening. As Fig. 1 shows, ^{196}Hg in natural mercury does not contribute much to the radiation because of its very low concentration (0.146 percent), nor do its emission and absorption heavily overlap with the other hyperfine components. Therefore, by increasing its concentration we expected to offer an additional channel for the 253.7-nm photons and thereby reduce average imprisonment time (τ_t) (normally about 10^{-5} second) and increase radiation efficiency.

We conducted a series of experiments with standard fluorescent lamps (34 and 40 W; diameter, 1.5 inches; length, 4 feet; normal operating temperature, 40°C). We monitored both ultraviolet output by Fabry-Perot interferometry as well as total light output in an integrating sphere as a function of total mercury pressure and ^{196}Hg concentration (*7*). As shown in Fig. 2, we obtained a maximum improvement in radiation efficiency of about 4.6 percent at ^{196}Hg concentrations of 2 to 4 percent.

At first, this maximum may appear to be somewhat puzzling. One intuitively would expect the maximum reduction in resonance trapping to be obtained when ^{196}Hg is present at a concentration comparable to that of the other five components (about 16 percent), to contribute a comparable line strength. However, there are two effects that reduce the ^{196}Hg concentration required for near optimum efficiency.

First, ^{196}Hg emission falls between the ^{198}Hg and combined ^{199}Hg and ^{201}Hg components (Fig. 3). A partial overlap of ^{196}Hg with the ^{201}Hg and ^{199}Hg components diminishes the gain in reduced resonance trapping time at higher ^{196}Hg concentrations.

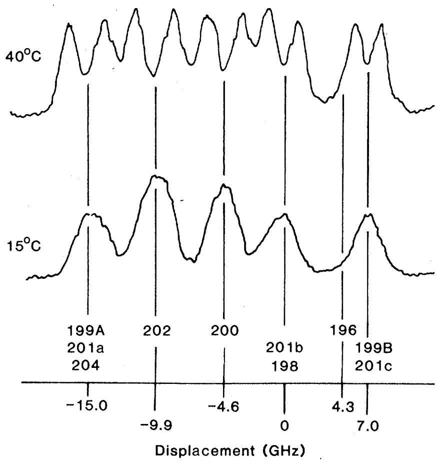
The second effect is the very strong resonance energy transfer between ^{196}Hg and all the other mercury isotopes. This process can be represented as

$$^{N}\mathrm{Hg}(6^3P_1) + {}^{196}\mathrm{Hg}(6^1S_0) \rightarrow {}^{N}\mathrm{Hg}(6^1S_0) + {}^{196}\mathrm{Hg}(6^3P_1) \quad (1)$$

with a cross section estimated at about 10^{-13} cm^2 (*8*) ; here 6^1S_0 and 6^3P_1 are the ground and first excited states of mercury, respectively. Since imprisonment time is proportional to the number density of the atoms and each isotope absorbs only its own radiation, a small concentration of ^{196}Hg offers an exit channel for the other hyperfine components through the energy transfer process of Eq. 1.

As the ^{196}Hg concentration increases, it becomes a less convenient exit channel because of its own increasing imprisonment time and the increasing importance of the back reaction in Eq. 1. A quantitative description of these effects can be obtained by solving a time-dependent differential equation for the imprisonment time of each isotopic component and summing the contribution of each.

Starting from Holstein's (*9*) original work and extending it to include a Voigt profile for the hyperfine components, hyperfine mixing by rare gases, energy transfer between the isotopes, and overlap of the components that make up the 253.7-nm line, we have developed a for-



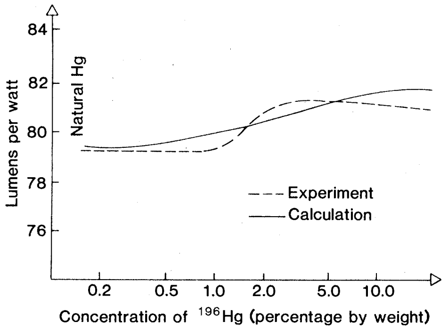


Fig. 1 (left). Fabry-Perot–measured hyperfine structure of the 253.7-nm resonance line of mercury in a mercury-argon low-pressure discharge of the type occurring in a standard fluorescent lamp at 15° and 40°C (*7*). Displacements are given in (*16*). Note the dramatic change in line shape between 15° and 40°C caused by the self-reversal phenomenon in resonance radiation (*17*). Fig. 2 (right). Experimental and calculated improvements in efficiency in a standard fluorescent lamp as a function of changes in the concentration of ^{196}Hg.

Table 1. Calculated reductions in resonance trapping time ($\tau_{rel} = \tau_t$ altered/τ_t natural) for various isotopic distributions. The "optimum" mixture is obtained by regression analysis. Except for mass numbers, values are percentages.

Mass number (amu)	Natural mercury	^{196}Hg 1 percent	^{196}Hg 3 percent	^{196}Hg 5 percent	Optimum
196	0.146	1.0	3.00	5.0	16.69
198	10.02	9.94	9.74	9.53	9.99
199	16.92	16.78	16.44	16.10	20.00
200	23.11	23.00	22.45	21.99	16.63
201	13.22	13.11	12.84	12.58	19.95
202	29.73	29.48	28.89	28.29	16.74
204	6.84	6.69	6.64	6.51	0.00
τ_{rel}	1.0	0.950	0.903	0.863	0.810

malism to calculate τ_t for any given isotopic composition. The differential equation for each isotopic component "i" is of the form

$$\frac{dn_i(\vec{r},t)}{dt} = -\frac{n_i(\vec{r},t)}{\tau_i} + \sum_k \frac{1}{4\pi\tau_k}$$

$$\int_0^\infty \epsilon_k(\nu)K_i(\nu)\,d\nu \int n_k(\vec{r},t)$$

$$\exp[-K(\nu)\,|\vec{r} - \vec{r}\,'|]/|\vec{r} - \vec{r}\,'d^3r' +$$

$$\sum_{k,i}[<\nu\sigma_{ki}(\nu)> N_i n_k - <\nu\sigma_{ik}(\nu)> n_i N_k]$$

$$+ N_g \sum_k (<q_{ki}\nu> n_k - <q_{ik}\nu> n_i) \quad (2)$$

where $n_i(\vec{r},t)$ and N_i are the number densities of the excited and ground states of the ith component, respectively; $\epsilon_k(\nu)$ is the frequency (ν)–dependent emission coefficient of the kth component; $k_i(\nu)$ is the absorption coefficient of the ith component; $K(\nu) = \sum_i K_i(\nu)$ is the total absorption coefficient; and $\vec{r}$ and t represent radial coordinate and time, respectively.

The first term on the right-hand side takes into account the decay of the excited state due to its natural lifetime τ_i; the second term represents the increase in $n_i(\vec{r},t)$ due to photon absorption; the third and fourth terms represent the change in $n_i(\vec{r},t)$ due to resonant energy exchange (cross sections σ_{ik}) and hyperfine mixing due to collisions (cross sections q_{ik}) with rare gases (density N_g).

Solution of this equation as a function of isotopic composition allows us to calculate τ_t of the 253.7-nm radiation. Introducing this reduced time into a complete discharge model, such as the one developed by Kenty (*1*) and Waymouth and Bitter (*10*), permits calculation of improvement in efficiency. As shown in Fig. 2, there is reasonably good agreement between experiments and theory (*11*). We should note that exclusion of the energy transfer, overlap, and mixing effects results in quantitative and qualitative disagreement with experiments.

A regression analysis was carried out to determine the isotopic composition that yields the minimum imprisonment time. The optimum composition (Table 1) is one that yields six hyperfine components of equal strength. Note the absence of ^{204}Hg; since ^{204}Hg emission overlaps almost completely with ^{199}Hg and ^{201}Hg, it does not contribute anything that is not already there. However, the reduction in τ_t with the optimum composition, which calls for 16 percent ^{196}Hg (~110-fold enrichment in ^{196}Hg, in addition to changes in the other isotopes) is only marginally better than that resulting from addition of 3 to 5 percent ^{196}Hg (20- to 34-fold enrichment). Therefore, it is likely that the economics for ^{196}Hg enrichment of only 3 to 5 percent would be more favorable than those for the optimum composition.

Accurate electrical and temperature measurements indicate that no significant changes would have to be made in the ballast, fixture, and lamp impedance design to accommodate this innovation.

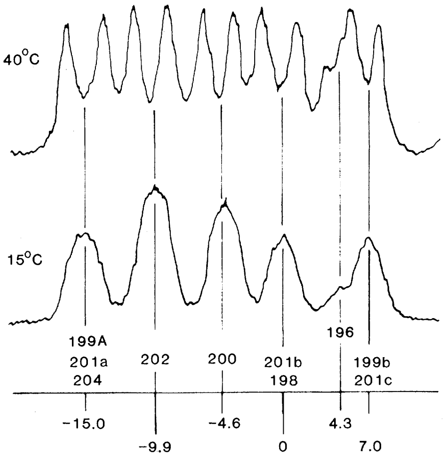


Fig. 3. Fabry-Perot–measured hyperfine structure of the 253.7-nm resonance line with 3.9 percent enriched ^{196}Hg in the same mercury-argon low-pressure discharge as in Fig. 1. Note the partial overlap of ^{196}Hg with ^{199}Hg and ^{201}Hg. As in Fig. 1, the change in line shape between 15° and 40°C is due to self-reversal (*17*).

Therefore, the improvements in efficiency could be implemented immediately, provided that natural mercury can be enriched in ^{196}Hg economically. Preliminary experiments in our laboratories to enrich ^{196}Hg by means of a photochemical reaction (*12, 13*) have given encouraging results (*14*). Some of the newer ^{235}U isotope separation techniques, such as atomic vapor laser isotope separation, the plasma separation process, or gas centrifuging, could conceivably be adapted to economically enrich mercury in ^{196}Hg.

Annual consumption of electrical energy for fluorescent lighting in the United States is about 1.8 × 10^{11} kWh (*15*). If 50 percent of the market converted to the new technology (a conservative estimate), savings, at 5¢ per kilowatt-hour, would total $200 million annually. As fluorescent lamps penetrate the residential market, the ultimate savings in energy costs may greatly exceed this figure.

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